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The dissolution kinetics of major elements in municipal solid waste incineration bottom ash particles

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Abstract

Leaching and tracer experiments in batches at L/S 20 were performed with 3-month-old MSWI bottom ash separated into eight different particle sizes. The time-dependent leaching of major elements (Ca^{2+}, K^+, Na^+, Cl^- and SO_4^{-2}) was monitored for up to 747 h. Physical properties of the particles, the specific surface (BET), pore volume and pore volume distribution over pore sizes (BJH) were determined for all particle classes by N_2 adsorption/desorption experiments. Some common features of physical pore structure for all particles were revealed. The specific surface and the particle pore volume were found to be negatively correlated with particle size, ranging from 3.2 m^2/g to 25.7 m^2/g for the surface area and from 0.0086 cm^3/g to 0.091 cm^3/g for the pore volume. Not surprisingly, the specific surface area was found to be the major material parameter that governed the leaching behavior for all elements (Ca^{2+}, K^+, Na^+, Cl^- and SO_4^{-2}) and particle sizes. The diffusion resistance was determined independently by separate tracer (tritium) experiments. Diffusion gave a significant contribution to the apparent leaching kinetics for all elements during the first 10–40 h (depending on the particle size) of leaching and surface reaction was the overall rate controlling mechanism at late times for all particle sizes. For Ca^{2+} and SO_4^{-2}, the coupled effect of diffusion resistance and the degree of undersaturation in the intra particle pore volume was found to be a major rate limiting dissolution mechanism for both early and late times. The solubility control in the intra particulate porosity may undermine any attempt to treat bottom ash by washing out the sulfate. Even for high liquid/solid ratios, the solubility in the intra-particular porosity will limit the release rate.

Keywords: kinetic; leaching; bottom ash; batch experiments

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1. Introduction

In order to successfully reuse municipal solid waste incineration bottom ash (MSWI BA) in civil engineering works without jeopardizing the goal of environmental protection, in particular the protection of groundwater quality, a detailed characterization is required of the governing chemical and physical processes that control the release of substances from MSWI BA under a variety of conditions as well as spatial and temporal scales.

The major elements in MSWI bottom ash are silicon, aluminum, iron and calcium. High concentrations of trace elements together with inorganic salts characterize the MSWI residues and are of major environmental

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concern. The release of trace metals and the subsequent transport process is primarily governed by pH. A number of alterations or aging processes initiate when the ash is exposed to water and atmosphere, which will contribute to the acid neutralization capacity (ANC) and govern the development of pH. The most important are the dissolution and precipitation of minerals of major cations, the slow weathering of glass phases and formation of clay-like materials and the formation of carbonates by uptake of carbon dioxide (Chandler et al., 1997). The pH level of MSWI bottom ash under alkaline and neutral conditions is mainly governed by the relatively fast dissolution of Ca and Mg minerals (Johnson et al., 1995; Yan et al., 1998). The dissolution process is highly time dependent, some reactions take place instantaneously while others, such as the weathering of the amorphous glass phases, are very slow and depend on the dissolution kinetics of the matrix (Yan et al., 1998). The dissolution of the inorganic salts is pH independent and relatively fast. Accordingly, the initial leachate from MSWI bottom ash contains high contents of chloride, sulphate, sodium, potassium and calcium (Hjelmar, 1996). With the exception of sulphate, which is solubility controlled and may precipitate as gypsum or ettringite (Kirby and Rimstidt, 1993; Meima and Comans, 1997; Dijkstra et al., 2006), the leaching of these substances are availability controlled.

At particle scale the dissolution process is constituted by a sequence of processes and includes transport of the reactant to mass transport boundary layer, diffusion through the boundary layer and micropores to the external and internal reaction surfaces, attachment on the surface, chemical surface reaction(s), detachment of the reaction products and finally the transport out into the bulk solution. The slowest process will govern the overall dissolution rate. According to the data on physical properties of ash particles, compiled by Chandler et al. (1997), the bottom ash particles are irregular and angular shaped, have a high specific surface area, a high pore area and small pore diameters compared to fly ash. Zevenberger et al. (1994) found that, although the smallest particles (<300 μm) constituted less than 35% of the total mass, more than 60% the total surface area was associated with this fraction. Bottom ash particles are also characterized by highly tortuous and long diffusion paths, dissolution of easily soluble minerals from large aggregates has therefore been hypothesized to be controlled by diffusion (Chandler et al., 1997; Yan et al., 1998).

This study addresses the governing processes for time-dependent leaching of inorganic salts from MSWI bottom ash in batch scale. Physical characterization together with leaching and tracer experiments in batches was performed with samples of eight different particle sizes. The main objective was to investigate the relative importance of diffusion control versus surface controlled reactions and degree of undersaturation for the overall leaching behavior of the major cations and anions Ca^{2+}, K^+, Na^+, Cl^- and SO_4^{-2}.

2. Model

2.1. Conceptualization

The surface reaction rate at the internal (and external) surfaces, the rate of diffusion from the particle pore space out in the bulk solution and the degree of undersaturation are important mechanisms that contribute to the apparent kinetics of the dissolution process in a batch system. The surface reactions is assumed to take place on all exposed surfaces of the particle. The experimental data indicate that a fraction of the mass dissolve instantaneously. This mass may originate from ultrareactive very fine particles (Berner, 1981; Strömberg and Banwart, 1999) and from easily dissolvable precipitates that cover the outer particle surfaces, perhaps as a result of the quenching process (bottom ash at several hundred degrees drop at the end of the furnace into a water lock before being dredged to the bottom ash bunker). The major cations and anions Ca^{2+}, K^+, Na^+, Cl^- and SO_4^{-2} are assumed to be released according to their availability with the exception of Ca^{2+} and SO_4^{-2}, where the release is assumed to be governed by the solubility of gypsum (used as model for the assemblage of minerals that may be controlling) although the saturation index of ettringite might be controlling at higher pH 10 (Dijkstra et al., 2006).

The ash particles are porous and irregularly shaped. In this study, the specific surface and the particle porosity was determined by BET N_2 experiments together with an estimation of the particle pore size distribution. However, a detailed description of the physical properties of the particles is not available, such as the spatial distribution of the particle pore space or the geometrical configuration of the pores. Assumptions of a homogeneous spatial distribution of porosity, reactive surfaces and chemical composition are hard to justify. Lacking this information, an approximate model for the diffusion process is employed here.

2.2. The approximate diffusion model

The spatial distribution of intra particle concentrations are here replaced by a particle volume-averaged concentration. The second order spatial derivate that appears in Fickian diffusion models is thereby eliminated. The rate of diffusion is here described as a linear function of the